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# **HIGH $T_c$ SUPERCONDUCTOR/ FERROELECTRIC HETEROSTRUCTURES**

**SRI International**

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## ABSTRACT

Thin films of the ferroelectric perovskite,  $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  (BST), were deposited on superconducting (100)  $\text{YBa}_2\text{Cu}_3\text{O}_x$  (YBCO) / (100) Ytria-stabilized zirconia (YSZ) substrates and (100) Si by ion-beam sputtering. Microstructural and compositional features of the ceramic bilayer were assessed by a combination of x-ray diffraction (XRD) and scanning electron microscopy. The films were smooth and featureless, and energy dispersive x-ray spectroscopy (EDX) data indicated that film composition closely matched target composition. XRD analysis showed that films deposited on YBCO substrates to be highly c-axis textured, while the films deposited on (100) Si did not exhibit any preferred growth morphology. The superconducting properties of the YBCO substrate layer were maintained throughout the processing stages, and as such, it was demonstrated that ion beam sputtering is a viable method for the deposition of Ferroelectric/YBCO heterostructures.

## INTRODUCTION

The research on thin film ferroelectric materials has been primarily driven, in recent times, by the development of the non-volatile ferroelectric random access memory (FRAM) device, and the reader is referred to the Reference literature list (particularly, the work of Ramesh et.al.) for details. In general, and similar to other applications of electronic ceramic thin films, the performance of FRAM devices is highly influenced by the crystalline quality of the ferroelectric film and the characteristics of the film-electrode interface. By utilizing a substrate (i.e., bottom electrode) which is both lattice and chemically matched, it should be possible to induce epitaxial growth of the ferroelectric layer and thus minimize defect structures associated with grain boundary effects. The perovskite ferroelectrics (e.g.,  $(\text{Ba},\text{Sr})\text{TiO}_3$ ,  $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ , etc.) and the perovskite YBCO superconductor possess similar lattice structure (i.e., 2-3% lattice match in the *a-b* plane) and crystal chemistry and thus provide an ideal system from which to base an experimental program. While the work of Ramesh et.al. at Bellcore has demonstrated that important ferroelectric performance properties (i.e., fatigue and ageing) of pulsed laser-deposited  $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$  were improved when the films were grown heteroepitaxially on YBCO, they did not comment on the superconducting properties of the YBCO electrode. An objective of this work was to investigate how the superconducting properties of the YBCO layer was effected by the deposition processing of the BST layer.

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## EXPERIMENTAL

### *Thin Film Processing*

$\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  ( $x = 0.25, 0.75$ ) films were ion beam sputter deposited from stoichiometric ceramic oxide targets. A 3 cm Kaufman source (Commonwealth Scientific, Inc.) was utilized to generate the  $\text{Ar}^+$  ion sputter beam which was positioned at an incident angle of  $45^\circ$  to the target. A low energy, gridless ion source (Commonwealth Scientific, Inc.), which was directed at the substrate with an incident angle of  $45^\circ$ , was utilized to preclean (by  $\text{Ar}^+$  ion sputtering) the substrates prior to film deposition. A gas dispersion ring located near the substrate was used to introduce oxygen during deposition. Films were deposited at a total pressure of  $2.0 \times 10^{-4}$  torr ( $1.0 \times 10^{-4}$  torr Ar and  $1.0 \times 10^{-4}$  torr  $\text{O}_2$ ). Substrate temperature was varied within the range of 550 to 650  $^\circ\text{C}$  over the course of this study. Deposition rate was on the order of 0.3 to 0.6 nm/min., and final film thickness was on the order of 200 to 250 nm as measured by surface profilometry.

The YBCO/YSZ substrates were commercially obtained (Excel Superconductor, Bohemia, NY) and were used as is. The silicon substrates were briefly etched in 10% aqueous HF and rinsed in distilled, deionized  $\text{H}_2\text{O}$  prior to entry to the vacuum system. Given that the principal study related to film deposition on YBCO and that the Si substrate depositions were only done for comparison purposes, it is noted that the film deposition conditions would in all likelihood result in the formation of an  $\text{SiO}_2$  layer on the Si which would negatively impact the quality of the BST/Si film samples. Prior to film deposition, the substrates were heated to the deposition temperature and sputter cleaned (i.e., removal of approximately a 10 nm surface layer) by a low-energy  $\text{Ar}^+$  ion beam.

## RESULTS and DISCUSSION

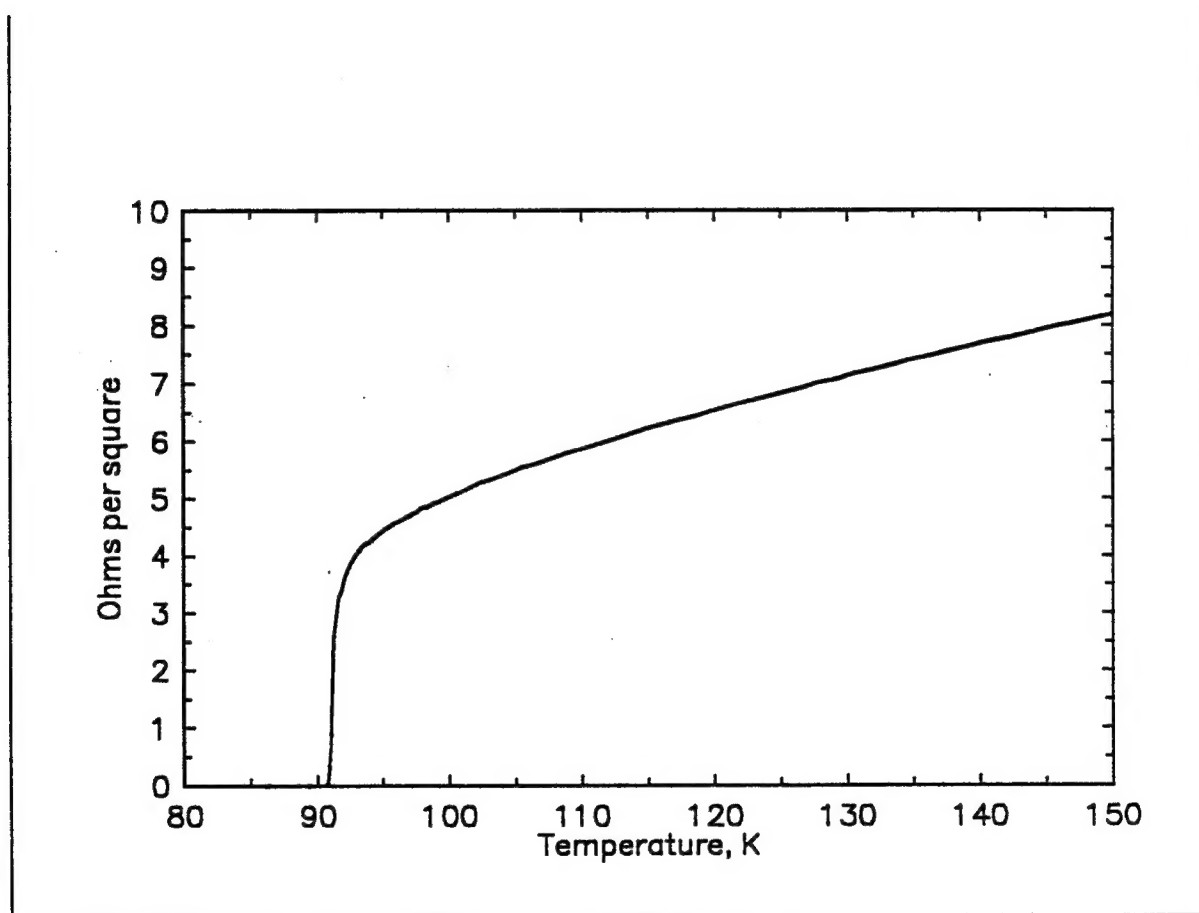
BST films deposited *in situ* at a substrate temperature of approximately 650  $^\circ\text{C}$  were visually observed to be smooth and optically reflective. Electrical measurements showed that the resistivity of the crystalline BST films ranged from  $10^4$  to  $10^8 \Omega\text{-cm}$ , with the more resistive films associated with higher deposition temperatures. Bulk sample data reported in the literature indicate a resistivity on the order of  $10^9$  to  $10^{10} \Omega\text{-cm}$  and as such, it is presumed that low level impurities (not detected by EDX measurements) and/or defect structures (i.e., oxygen vacancies) reduced film resistivities. Due to the high leakage current of the deposited films, the ferroelectric properties of a test Pt/BST/YBCO capacitor structure could not be analyzed.

XRD analysis indicated that films deposited on (100) YBCO were highly c-axis oriented, while films deposited on (100) Si did not exhibit any preferred orientation effect. This is

consistent with our previous results relating to the sol-gel processing of  $\text{Pb}(\text{Zr,Ti})\text{O}_3$  films on (100) YBCO and Pt-coated Si, but the microstructural quality of the sputtered films were superior to that of the sol-gel-derived films.

The superconducting properties of the YBCO base layer were determined by the DC four probe method both prior and after deposition of the BST overlayer. Figure 1 shows that *in situ* BST film processing had minimal effect on the electrical properties of the YBCO layer as a

Figure 1



sharply-defined  $T_c$  of approximately 90 K is maintained.

## CONCLUSIONS

$\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  ( $x=0.25, 0.75$ ) films were deposited *in situ* on (100) YBCO/YSZ substrates by ion beam sputtering. Based on EDX results, and substantiated by the analysis of XRD data, the composition of a deposited film was consistent with the stoichiometry of the single oxide target utilized. XRD analysis showed that films deposited on "lattice-matched" YBCO substrates to be highly c-axis textured, while films deposited at the same growth conditions on (100) Si did not exhibit any preferred crystallographic orientation. The resistivity of the ion-beam sputtered BST films were lower than bulk reported values and is probably related to low level impurities and/or oxygen defects. The superconducting properties of the YBCO base layer were not effected by the BST deposition process, and as such ion beam sputtering affords a viable method for the fabrication of Ferroelectric/Superconductor -based device structures.

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